Kondo effect in double quantum dots with inter-dot repulsion

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We investigate a symmetrical double quantum dot system serially attached to the leads. The emphasis is on the numerical analysis of finite inter-dot tunneling in the presence of inter-dot repulsive capacitive coupling. The results reveal the competition between extended Kondo phases and local singlet phases in spin and charge degrees of freedom. The corresponding phase diagram is determined quantitatively.

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Quantum dots[1,2,3,4] provide in addition to various applications in proposed spintronics[5] and quantum computation[6,7] devices a playground for studying phenomena known in bulk condensed-matter systems. Specifically, the Kondo effect was found to play an important role in single[8] and double quantum dot[9,10,11] (DQD) systems. From early theoretical work on the low-temperature properties of the two-impurity Kondo Hamiltonian[13,14] it is known that localized moments form either each its own Kondo singlet with delocalized electrons in the leads – *double Kondo* phase (2K) – or they form *local spin-singlet* state (LSS) decoupled from delocalized electrons. The crossover between the two regimes is the consequence of competing energies of Kondo singlet with Kondo temperature T_K and of LSS formation J. Similar results were obtained by the analysis of a two-impurity Anderson model by means of slave-boson formalism[15,16,17,18,19] and numerical renormalization group (NRG)[20,21]. Resembling behavior was found also in particular regimes of a triple quantum dot system[22].

Here we focus on the role the inter-dot (capacitive) interaction plays in the electron transport through serially coupled DQD. The influence of the capacitive coupling was already studied by means of the equation-of-motion method[23,24,25,26] which, however, fails to capture the Kondo correlations accurately. On the other hand, the Kondo correlations were considered in the limit of vanishing tunnel coupling and strong inter-dot interaction where the ground state of the isolated DQD exhibits four-fold degeneracy: in addition to spin degeneracy also singly occupied states labeled with $(1,0)$ and $(0,1)$ are degenerate leading to orbital Kondo behavior[27,28,29,30,31]. Here (n_1, n_2) corresponds to occupancies n_1 and n_2 of the two dots. The simultaneous presence of spin and orbital degeneracy results in enhanced Kondo temperature $T_K(N) \sim \sqrt{J'\rho_0 N}\exp(-1/J'\rho_0 N)$, where N is the degeneracy, ρ_0 the noninteracting density of states, and J' the corresponding Schrieffer-Wolf prefactor[28]. The isospin Kondo resonance was observed indirectly in the measurements of enhanced conductance through DQDs[9,10,11,12] and carbon nanotubes[32,33] and also directly, in the bulk, with techniques of the scanning tunneling microscopy[34]. Recently the *enhanced Kondo temperature* phase (EKT) was predicted for DQD with inter-dot interaction $V = U$ in the absence of tunneling between the dots also at half-filling[35], i.e., $n = n_1 + n_2 = 2$. The main characteristics of this phase at low temperatures is the enhanced width of the Kondo resonance (i.e., Kondo temperature T_K) on top of an incoherent continuum in the density of states. However, the analysis of capacitively coupled DQD with finite inter-dot tunneling rates has been lacking to date and we address this issue in the present paper.

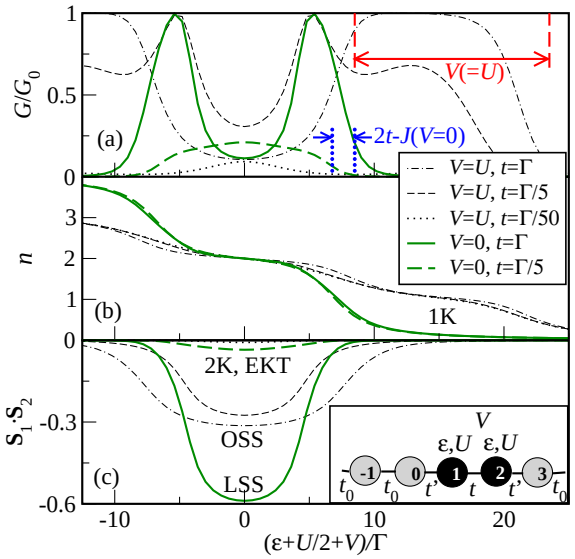

Figure 1: (Color online) Conductance (a), occupancy (b) and spin-spin correlation (c) of DQD for $V = 0$ (thick), and $V = U$ (thin). The vertical lines for $V = U$ (dashed), and $V = 0$ (dotted) correspond to transitions of isolated DQD between $n = 0, n = 1$ and $n = 1, n = 2$ for $t = \Gamma$. Spin-singlet regimes (LSS, OSS) and Kondo regimes (2K, EKT, 1K) are indicated with labels. Inset: DQD system with both: inter- and intra-dot Coulomb coupling, attached to noninteracting leads.

We model DQD shown in inset of Fig. 1(c) by the Anderson-type Hamiltonian $H = H_{\rm d} + H_{\rm l}$, where $H_{\rm d}$ corresponds to the isolated dots

$$H_{\rm d} = \sum_{i=1,2} (\epsilon n_i + U n_{i\uparrow} n_{i\downarrow}) + V n_1 n_2 - t \sum_{\sigma} (c^\dagger_{1\sigma} c_{2\sigma} + h.c.),$$

with $n_i = n_{i\uparrow} + n_{i\downarrow}$, $n_{i\sigma} = c^\dagger_{i\sigma} c_{i\sigma}$. The dots are coupled by a tunneling matrix element t and a capacitive V term. The on-site energies ϵ and the Hubbard repulsion U are taken equal for both dots. $H_{\rm l}$ corresponds to the noninteracting left and right tight-binding lead and to the coupling of DQD to the

leads,

$$H_{\rm l} = -t_0 \sum_{i\neq 0,1,2} c^\dagger_{i\sigma} c_{i+1\sigma} - t' \sum_\sigma (c^\dagger_{0\sigma} c_{1\sigma} + c^\dagger_{3\sigma} c_{2\sigma}) + h.c.,$$

where the sites are labeled as shown in the inset of Fig. 1(c).

In this paper we study the low temperature properties of DQD, derived from the ground state at $T = 0$, determined by the Gunnarsson and Schönhammer projection-operator method[36,37]. The conductance is calculated using the sine formula[37], $G = G_0 \sin^2[(E_+ - E_-)/4t_0 L]$, where $G_0 = 2e^2/h$ and $E_\pm$ are the ground state energies of a large auxiliary ring consisting of L non-interacting sites and an embedded DQD, with periodic and anti-periodic boundary conditions, respectively. The chemical potential is set in the middle of the band, which corresponds to L electrons in the ring. The method proved to be very efficient for various systems with Kondo correlations[22,37,38,39]. Up to $N \sim 5000$ sites were used with variational ansatz based on ~ 100 projection operators, which was sufficient to obtain converged results for the boundaries between various regimes.

For large inter-dot tunneling $t \gtrsim U$ molecular bonding and anti-bonding orbitals with energies $\epsilon_{b(a)} = \epsilon \mp t$ are formed. Whenever either of the two orbitals is singly occupied, the physics is analogous to the single impurity Anderson problem with unitary transmission and Kondo correlations (1K regime). When the bonding orbital is doubly occupied the electrons form an *orbital spin-singlet* state (OSS) with diminished conductance as the Kondo peak in the density of states is absent. The other limit, $t \to 0$, is more interesting. When also inter-dot repulsion is moderate $V \lesssim U$ the dots are effectively decoupled and in the particle-hole symmetric point ($\epsilon + U/2 + V = 0$) the 2K state occurs. Regimes with different occupancies are separated by the intermediate valence regime of width determined by the noninteracting hybridization $\Gamma = t'^2/t_0$.

Behavior of the system is less obvious in the intermediate region $0 \lesssim t \lesssim U$. In Fig. 1(a-c) we plot the conductance, occupancy and spin-spin correlation for $V = 0$ and $V = U$ ($\Gamma = t_0/25$ and $U/\Gamma = 15$ is kept constant throughout the paper). The curves for $t = \Gamma, V = U$ (dashed-dotted thin lines) are the most reminiscent of known results for single impurity case. The wide plateau in conductance curve corresponds to single occupancy regime. While the occupancy for the case with reduced $t = \Gamma/5$ (dashed thin line) does not change significantly, two maxima in conductance become discernible. One reflects the 1K Kondo physics corresponding to the bonding orbital ($n = 1$ there). The other maximum (with unitary conductance) is known from the $V = 0$ case[16,21] and occurs generally at the transition between extended Kondo and decoupled spin-singlet states at $n \sim 2$. For smaller $t = \Gamma/50$ at $n \sim 1$ conductance is significantly reduced, but the ground state is still 1K, which is signaled with the remaining plateau in occupancy and diminished fluctuations of the occupation of the relevant, (*i.e.*, bonding or anti-bonding) orbital (not shown here). In this case the OSS state is not energetically favorable near $n \sim 2$, correspondingly the conductance peak with the unitary conductance is absent (likewise for $V = 0$, $t = \Gamma/5$, where the LSS is not energetically preferred). At $n = 2$ rather

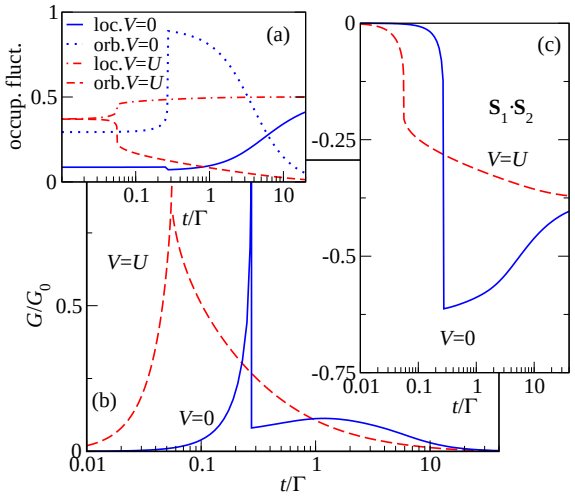

Figure 2: (Color online) (a) Orbital $\Delta n_b^2 = \Delta n_a^2$ and site $\Delta n_1^2 = \Delta n_2^2$ occupancy fluctuations at half-filling ($n = 2$) as a function of t/Γ for $V = 0$ and $V = U$. Conductance (b) and spin-spin correlation (c), for $V = 0$ (full) and $V = U$ (dashed).

the EKT phase is found, as discussed later.

The $V = 0$ case is seen to differ qualitatively from the $V = U$ case. Crude insight into distinction between the two may be gained from treating just the filling properties of a detached system[40]. The first electron is added when $\epsilon = t$, and the second when $\epsilon = -t + J + [(U+V) - |U-V|]/2$, where $J = [-|U-V| + \sqrt{(U-V)^2 + 16t^2}]/2$ is the difference between singlet and triplet energy of isolated DQD. When $n = 2$ the ground state is $[\alpha(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle) + \beta(|20\rangle - |02\rangle)]/\sqrt{2}$, where $\alpha/\beta = 4t/(V - U + \sqrt{(U-V)^2 + 16t^2})$. For $V = 0$ and $V = U$ case the values of ϵ where the occupancy of a ground state changes from $n = 0$ to $n = 1$ and from $n = 1$ to $n = 2$ are indicated by vertical lines on Fig. 1(a). The range of ϵ where single occupation is favorable is diminished in the $V = 0$ case.

This observation remains valid also for DQD attached to the leads, Fig. 1(b). At first sight surprising feature is seen in Fig. 1(c), where the expectation value of a spin-spin correlator $\mathbf{S}_1 \cdot \mathbf{S}_2$ is seen to approach $-3/8$ near $n = 2$, but in fact it is just equal to the noninteracting result with two electrons occupying the bonding orbital. The fact that the spin-spin correlator approaches $-3/4$ near $n = 2$ in the $V = 0, t = \Gamma$ case suggests that the local picture prevails. The electrons indeed form singlet in the local basis what is seen also in diminished value of the local occupancy fluctuations, Fig. 2(a). Additional insight into precise role of inter-dot interaction can be obtained by re-writing the total Hamiltonian in the basis of orbital operators,

$$H_{\rm d} = \sum_{\alpha=a,b} \left[\epsilon_\alpha n_\alpha + \frac{U+V}{2} (n_{\alpha\uparrow} n_{\alpha\downarrow} + n_{\alpha\uparrow} n_{\bar{\alpha}\downarrow}) \right] + \\ + V \sum_\sigma n_{a\sigma} n_{b\sigma} + \frac{U-V}{2} (C_{\rm flip} - S_{\rm flip}),$$

where notation $\bar{a} = b$, $\bar{b} = a$ is used. The last term of $H_{\rm d}$ consists of charge-flip $C_{\rm flip} = T_a^+ T_b^- + h.c.$ and spin-flip $S_{\rm flip} = S_a^+ S_b^- + h.c.$ operators, where $S_\lambda^- = c_{\lambda\downarrow}^\dagger c_{\lambda\uparrow} = (S_\lambda^+)^\dagger$ are spin and $T_\lambda^- = c_{\lambda\uparrow} c_{\lambda\downarrow} = (T_\lambda^+)^\dagger$ charge (isospin)[41] lowering and raising operators for the orbitals $\lambda = b, a$ (or sites $\lambda = 1, 2$). The full spin (isospin) algebra is closed with operators $S_\alpha^z = (n_{\alpha\uparrow} - n_{\alpha\downarrow})/2$ and $T_\alpha^z = (n_\alpha - 1)/2$, respectively.

When $V = U$, the spin- and isospin-flip terms in $H_{\rm d}$ are absent: the Hamiltonian is mapped exactly to the two-level Hamiltonian with intra- and inter-level interaction U with the bonding and anti-bonding levels coupled to even and odd transmission channels, respectively. When $V \neq U$ this mapping is no longer strictly valid. Taking $V = 0$ case as an extreme example, we find two mechanisms responsible for this. Firstly, the electrons can avoid the inter-level repulsion by occupying aligned spin-states in different orbitals, and secondly, charge flip-terms induce the fluctuations of charge between orbitals. Both mechanisms prohibit electrons to occupy the well-defined orbital states.

In the rest of the paper we concentrate on the symmetric point of the model, $n = 2$. The manifestation of adequacy of orbital picture are the fluctuations of occupancy $\Delta n_\lambda^2 = (n_\lambda - \langle n_\lambda \rangle)^2$, shown in Fig. 2(a). For $V = U$ orbital occupancy fluctuations Δn_b^2(= Δn_a^2 for $n = 2$) are indeed smaller than local occupancy fluctuations $\Delta n_1^2 = \Delta n_2^2$ for all values of t. On contrary, for $V = 0$, $\Delta n_b^2 < \Delta n_1^2$ only for $t \gtrsim 5U$.

To explain the conductance and spin-spin correlation shown in Fig. 2(b,c) we first note that the electrons bind into singlet (local or orbital, depending on the V/U), whenever $J \gtrsim J_c = 2.2T_K$, which is known for $V = 0$ results[13,43], but we find that this result can be readily generalized for by taking the difference between singlet and triplet energies J also for $V > 0$. This relation indicates that although the Kondo temperature is known to rise in the $V = U$ case[35] for $n = 2$, the OSS is energetically favorable for moderate t as $J(V \sim U) \propto t$ is larger when compared to $J(V = 0) \sim 4t^2/U$.

It should be noted that due to the variational nature of the method, the boundaries between various regimes are accurately reproduced. Sharp transitions of correlation functions in the vicinity of crossovers are less precisely determined. The transitions discussed here are actually smoothened into crossovers what happens generally when the parity breaking terms (here the inter-dot tunneling) are present in the Hamiltonian[42].

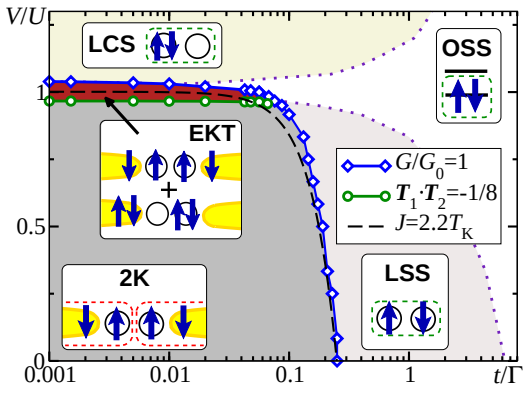

Figure 3: (Color online) Phase diagram in $(t/\Gamma, V/U)$ plane for $n = 2$.

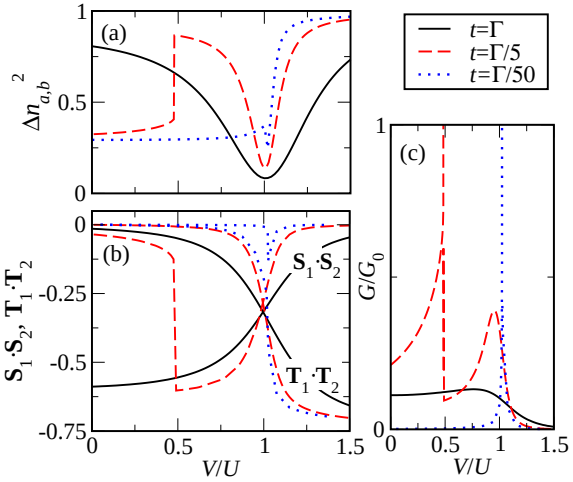

Figure 4: (Color online) (a) Fluctuations of charge in orbitals $\Delta n_a^2 = \Delta n_b^2$, (b) the spin-spin $\mathbf{S}_1 \cdot \mathbf{S}_2$ and isospin-isospin correlation $\mathbf{T}_1 \cdot \mathbf{T}_2$ are plotted for $t > t_c$ (full), for $t_c^* < t < t_c$ (dashed) and $t < t_c^*$ (dotted). (c) The conductance for the three cases.

For $V = 0$ in absence of inter-dot tunneling $t \to 0$, each of the dots forms a Kondo singlet with its own lead (2K regime). When t is increased above some value t_c, set by $J \gtrsim J_c$, the ground-state becomes a LSS, what is seen from increased value of $-\mathbf{S}_1 \cdot \mathbf{S}_2 \to 3/4$. The conductance is small in both cases due to the effectively decoupled leads in the 2K case or deficiency of states near the chemical potential in the LSS case. The conductance $G \to G_0$ at some point what is most vividly understood in terms of the phase shift between odd and even transmission channels, changing from 0 to π during the transition[16]. For $V \lesssim U$ the behavior does not change qualitatively, as the transition remains close to $2T_K \sim J$. In the case $V \sim U$ numerical results show deviations from this prediction: the transition occurs at smaller values of t than set with relation $J \gtrsim J_c$. We explain this by increased Kondo temperature in the EKT phase as suggested from NRG results[35]. Here the transition between 'SU(4) Kondo'[44] and OSS occurs at $T_K^{SU(4)} \sim J(V = U) = 2t_c^*$.

Further increasing V above U we find, that in contrast to the strict $t = 0$ case where the ground state for $V \gtrsim U$ is a non-Fermi liquid charge-degenerate phase[35], a finite t breaks the degeneracy resulting in a (Fermi liquid) phase with increased magnitude of isospin-isospin correlation $\mathbf{T}_1 \cdot \mathbf{T}_2$: a *local charge singlet* (LCS). LCS corresponds to the ground state of a detached system for $(V - U)/t \gg 1$.

In Fig. 3 the phase diagram of DQD for $n = 2$ is shown. The border line between Kondo and local-singlet phases (full line with diamonds) is characterized by unitary conductance and the abrupt increase of $-\mathbf{S}_1 \cdot \mathbf{S}_2$ for the transition between 2K and LSS and of $-\mathbf{T}_1 \cdot \mathbf{T}_2$ for transition between EKT and LCS [see also Fig. 2 (a,b) and Fig. 4(b,c)]. In the $t, t' \to 0$ limit the states $(2,0), (0,2)$ and four states $(1,1)$ are degenerate, and $\langle -\mathbf{T}_1 \cdot \mathbf{T}_2 \rangle = 1/6$, if the expectation value is taken

with respect to the ground state where each of these states occurs with equal probability. We quantitatively characterize the transition to the EKT phase (full line with circles) when $-\mathbf{T}_1 \cdot \mathbf{T}_2 > 1/8$. The border lines which we use to distinguish between local and orbital regimes (dotted) are given by $\Delta n_b^2 = \Delta n_1^2$ for $V < U$ and by $-\mathbf{S}_1 \cdot \mathbf{S}_2 = 3/16$ (half of noninteracting value) for $V > U$.

In Fig. 4 we sample the phase diagram at different t by increasing V. We plot three different cases. In the first case (full curve) DQD is in LSS already at $V = 0$. By increasing V we see a crossover to LCS through the orbital regime. In the other two cases we start in 2K. Now if $t < t_c^*$ (dotted) we stay in the Kondo regime until the transition to LCS happens with the unitary conductance at the transition point. However if $t > t_c$ (dashed) there is an area in between where LSS is energetically favorable. From the point where the system is in LSS, it follows the same scenario as in the first case.

Finally, let us briefly comment on $V > U$ part of the phase-diagram. This regime could arise when the dots are capacitively coupled while the on-site repulsion is reduced due to the coupling to local Einstein vibrational modes[39,45]. Here the competition between LCS and EKT is relevant for the ground-state. Our numerical data confirms NRG predictions that the Kondo regime is relevant only for small increase of V over U. For parameters used here, we find critical $V_c \sim U + U/20$.

In conclusion, we have presented the phase diagram of a pair of tunneling-coupled quantum dots with Coulomb inter- and intra-dot interaction. In general, the behavior of $V \sim U$ case was found to differ from the $V \sim 0$ case due to different arrangement of orbital levels of a decoupled system. In addition, for $V \sim U$ the correct exchange coupling is not $J \sim 4t^2/U$ but rather $J \sim 2t$, hence the spin-singlet binding energy is increased. The increase in J is seen to surmount the enhancement of Kondo temperature which is due to the additional degeneracy in the charge sector. Therefore this enhanced Kondo effect for $n = 2$ should be discernible in measurements only in a tiny window of tunneling rates and inter-dot vs. intra-dot interaction.

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